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# RPPR Final Report

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**Major Goals:** The main goals of this proposal are to experimentally realize novel topological insulator (TI)-based magnetic/antiferromagnetic heterostructures with comprehensive studies on their quantum physical properties. The interplay among band topology and symmetry and magnetism gives rise to more novel physics in such heterostructures. By interfacing the TI with some ferromagnets and antiferromagnets (AFM), the interfacial magnetic exchange can break the time-reversal-symmetry on the surface of the TI via exchange coupling or proximity effect. In this report, we focus on new types of TI/AFM heterostructures and superlattices. Via structural engineering and molecular beam epitaxy, both composition and thickness of TI and AFM layers can be accurately controlled to provide a versatile experimental platform for exploring the exchange interaction between the two material systems. The present study may result in possible new effects and various breakthroughs as well as applications, for example, the giant SOT in TIs and ultrafast resonance frequency in AFM will help to realize various low power-consumption spintronic devices, such as RF/THz oscillators based on spin-wave and super-spin currents. This work will also generate the necessary knowledge and skills to enable the next-generation of information process technologies.

**Accomplishments:** In this report, we achieved high quality AFM/MTI heterostructures and superlattices using molecular beam epitaxy method. The AFM material CrSb and MTI can form high quality interfaces to facilitate our study. We successfully realized emergent interfacial magnetic effects through the exchange coupling between topological Dirac Fermions and AFM order mediated by AFM spins. By using the state-of-art polarized neutron reflectometry technique with collaboration with NIST, we revealed origin of the interfacial magnetic coupling effect between MTI's ferromagnetic order and CrSb's AFM order. The proximity effect between the two material systems raises the Curie temperature of MTI by three times which is critical for potential topological-insulator-based device applications. Beyond the magnetic interaction, we also reveal a possible topological phase transition in the AFM/TI/AFM trilayer structure which may potentially serve as a platform for axion insulator study. Furthermore, the effective coupling between massive Dirac fermions with the antiferromagnetic order creates enormous opportunities for integrating and modifying topological surface states, and opens up new avenues towards topological antiferromagnetic spintronics.

**Training Opportunities:** Nothing to Report

**Results Dissemination:** The results described in this report were published on Nature Materials, 16 (1): 94-100. "Tailoring Exchange Couplings in Magnetic Topological-Insulator/Antiferromagnet Heterostructures" and Physical Review Letters 121, 096802 "Topological Transitions Induced by Antiferromagnetism in a Thin-Film Topological Insulator"

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**Participant:** Kang Wang

**Person Months Worked:** 12.00

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**Participant:** Yabin Fan

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**Authors:** YABIN FAN, QIMING SHAO, XUFENG KOU, PRAMEY UPADHYAYA, KANG WANG

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**Authors:** Yabin Fan

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# Scientific progress and accomplishments

## Foreword

Recently, introducing ferromagnetism to break the time-reversal-symmetry in topological insulator has raised great interest to create massive Dirac fermions. Such process can be done through either magnetic element doping into the system or through proximity effect by interfacing topological insulator with another ferromagnetic or antiferromagnetic materials [1-4]. As a result of the topological protection, the Dirac surface states show up at the surface of TI and exhibit various exotic physical properties, including the spin-momentum locking, chiral conduction protected by time reversal-symmetry, spin-orbit torque (SOT), axion electrodynamics, etc. One of the most demanding effect is the quantum anomalous Hall effect by doping Cr or V into the  $(\text{BiSb})_2\text{Te}_3$  system where dissipationless edge state transport shows up without the assistance of external magnetic field [5-9]. Also, by interfacing TI with MTI, giant SOT shows up in the system that is almost three orders magnitude greater than heavy metal case [10, 11]. Introducing heterostructures, in which the SOC and the magnetic exchange can be manipulated individually, may not only further improve these properties but also make possible to uncover other new ones. New interfaces combining magnetic material with TI may effectively break the time reversal symmetry while at the same time introducing new magnetic responses into the system. Such structures may establish new research fields in condensed matter physics to understand the interplay between topology and magnetism. New physics may inspire a wide range of application-oriented ideas in ultra-low power electronics, quantum computing, spintronics and terahertz devices.

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## Statement of the problem studied

The research program focuses on the growth, device nanofabrication, physical property characterization and understanding the interface effects inside magnetic topological insulator and antiferromagnet heterostructures. By achieving deeper understanding in these structures, we are able to learn more effective methods in manipulating the massive Dirac fermion based magnetic structures and construct energy efficient topological spintronic devices and creates new research avenue in terms of MTI/AFM exchange coupling phenomena. To achieve this, we are approaching the structure in three major ways: Firstly, we are studying the transport property of the structure which provides us direct quantum transport behavior. Secondly, we are studying the overall magnetization behavior which gives us better understanding in the macroscopic improvement in MTI's ferromagnetism and AFM layer's coupling to it. Thirdly, we are studying the structure using polarized neutron reflectometry technique which probes the interface effect and reveals the in-depth magnetization profile in the most straightforward way. Combining the three techniques, we are trying to understand the spin-orbit coupling related physics in MTI when interfaced with AFM including the long range magnetic coupling throughout the whole heterostructure/superlattice, the macroscopic magnetization enhancement, the detailed magnetization profile and coupling mechanism and topological related behavior.

## Summary of the most important results

- (1) Successful integration of antiferromagnet CrSb with TI/MTI heterostructure superlattice using molecular beam epitaxy technique

We first successfully prepared high-quality heterostructures/superlattices consisting of an AFM, CrSb and MTI, Cr-doped  $(\text{BiSb})_2\text{Te}_3$  or undoped TI using molecular beam epitaxy. As shown in Fig. 1, the transmission electron microscopy image reveals the sharp interface between the two materials owing to the optimized growth condition. The two material systems possess similar lattice constant. And most critically, both material systems are in hexagonal symmetry along the c-axis which is essential to achieve high-quality heterostructure/superlattices. Such structure enables clean study of the interface effect and greatly reduces possible defects related physics or inter-diffusion related doping effect.

- (2) Multi-switching states in AFM/MTI heterostructure/superlattice

As shown in Fig. 2, the magnetization hysteresis loop develops multiple switching states when AFM is inserted in between MTI layers and such behavior becomes more pronounced with the increasing number of superlattice periods. Such feature is potentially as a result of the effective long-range exchange coupling, i.e., coupling between two neighboring MTI layers. We further investigated this phenomenon using quantitative Landau-Lifshitz-Gilbert simulation shown in Fig. 3 which reveals a new type of ferromagnetism interplay that causes such multi-switching states. Such interplay which involves massive Dirac fermions is very appealing to topological phenomena which requires intricate control of system's ferromagnetism. To directly probe this effect, we later used polarized neutron reflectometry to confirm it. It is found that the AFM layer serves as a coupling mediator with a modified spin texture that magnetically linked its top and bottom neighboring MTIs. Such effect is further confirmed by neutron technique as shown in Fig. 4.

- (3) Enhanced ferromagnetism in AFM/MTI heterostructure/superlattice



The third major finding is the enhanced ferromagnetism in MTIs utilizing this structure. The current MTI system which uses Cr as the doping element has a Curie temperature around 30 K. Further increasing the doping level will possibly damage the material's topological property which is unwanted. By interfacing high Néel temperature AFM ( $T_N \sim 700$  K) with MTIs, the Curie temperature of the Cr-doped MTI is enhanced by three times from 30 K to 90 K which brings any possible device application from liquid helium temperature to liquid nitrogen temperature. In the meantime, the coercive field increased by an order of magnitude shown in Fig. 5. Such enhancement opens up new opportunities in searching for high temperature topological phenomena utilizing AFM-based proximity effect.

#### (4) Topological phase transition when sandwiching undoped TI with two AFM

When undoped TI is sandwiched between two AFM layers, not only ferromagnetism shows up as a result of AFM induced proximity effect, but also a new type macroscopic transport behavior was observed. In the longitudinal resistance channel, a unique anti-symmetric peak-dip behavior shows up at the coercive field as shown in Fig. 6. Such behavior is not seen in MTI related structure or bilayer structure. Further study shows that during the magnetization reversal, intermediate spin configurations are ascribed from unsynchronized magnetic switching. This unsynchronized switching develops antisymmetric magnetoresistance spikes during magnetization reversals, which might originate from a series of topological transitions. This topological transition behavior might provide a new platform to study axion insulator related behaviors.

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## Appendixes

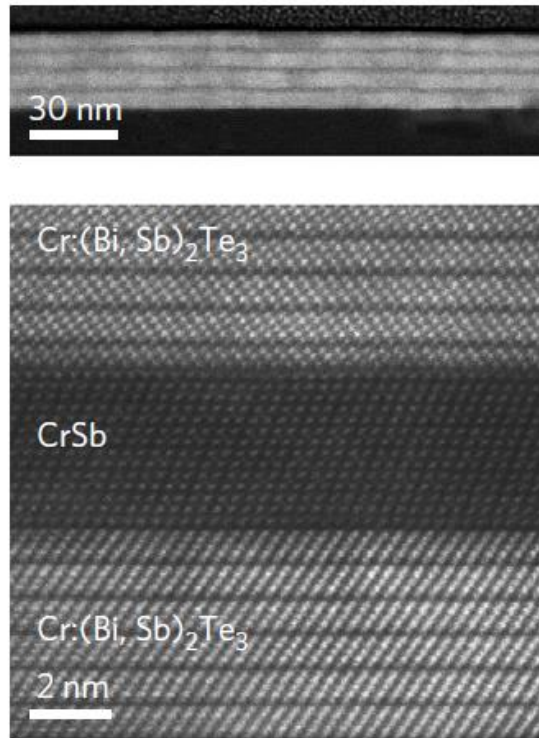


Figure 1 High-resolution transmission electron microscopy images of an AFM(CrSb)/MTI [Cr-doped  $(\text{Bi,Sb})_2\text{Te}_3$ ] superlattice.

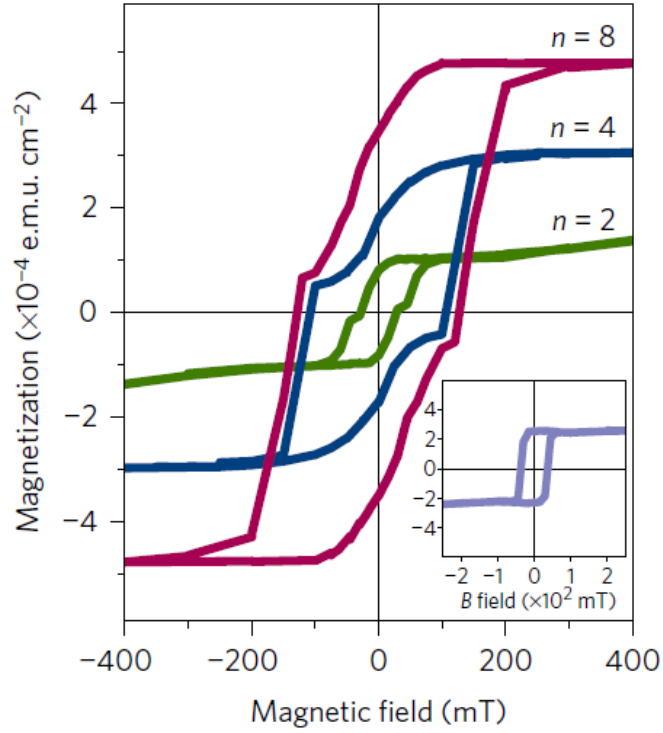


Figure 2 Antiparallel effective long-range exchange coupling in (AFM/MTI) $n$  SLs with  $n=2, 4$  and  $8$ , as demonstrated by the double-step behaviours in the M–H loops measured at  $5$  K. Similar to the trilayer in Fig. 1g, this behaviour is attributed to the antiparallel magnetizations between the neighbouring MTI layers in the SLs, which originates from the interactions between the MTI surface spins and AFM spins. The inset shows the magnetization of a control SL (TI/MTI) $n=4$ , in which the AFM layers are replaced by an undoped TI layer with the same thickness. In this case, the lack of exchange couplings between the Dirac fermions and AFM spins only contributes a single step switching behaviour.

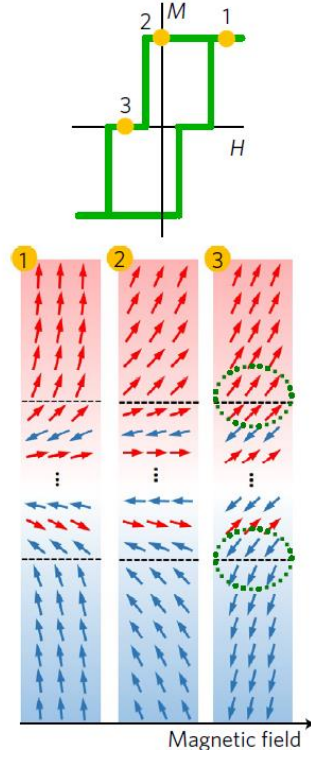


Figure 3 The relative orientations of the atomic moments of the effective long-range exchange coupling in a MTI/AFM/MTI trilayer, which is generated from a LLG quantitative simulation and calculation using the saturation magnetization obtained from the PNR results. The interfacial exchange coupling between the Dirac fermions and the AFM spins orients the magnetization directions of the two MTI layers, forming a stable field-induced Néel-type domain wall within the AFM.

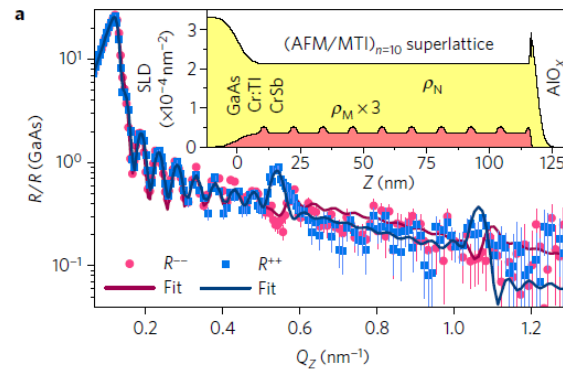


Figure 4 Polarized neutron reflectivities (at 20 K with a 700mT in-plane field) normalized to the GaAs substrates for the spin-polarized R++ and R-- channels of an (AFM/MTI) $n=10$ . The insets show the

corresponding models with structural and magnetic scattering length densities (SLDs), used to obtain the best fits, which contain magnetized TI and AFM layers for the SL, and a magnetized TI and two barely magnetized AFM layers for the trilayer. In the SL, the weak magnetization within the AFM layers is probably due to spin texture modification induced by proximity.

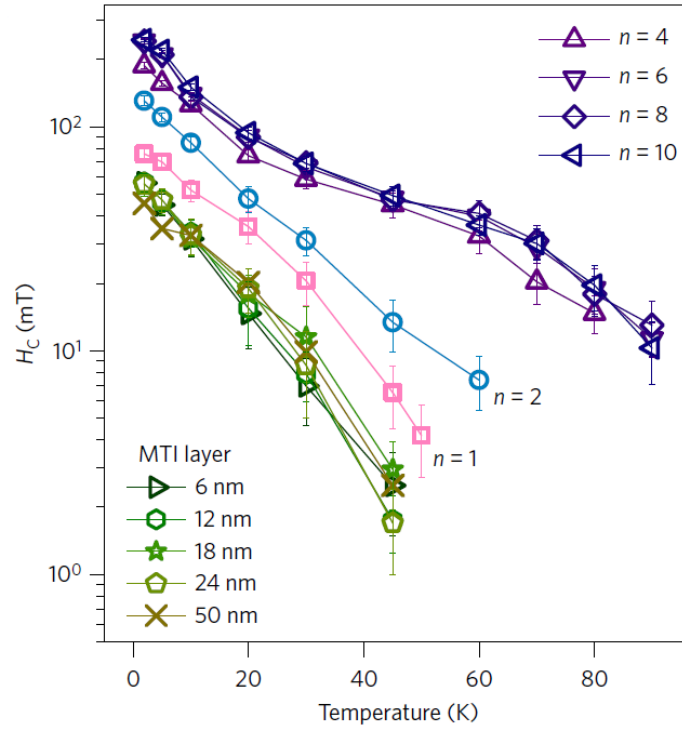


Figure 5  $H_C$  of MTI single layers and (AFM/MTI) $_n$  SLs versus  $T$ , showing  $H_C$  increases along with the increase of  $n$ , consistent with correlation between the effective long-range exchange coupling and magnetic ordering enhancement. The error bars specify the standard deviations of the measurements from the superconducting quantum interference device system.

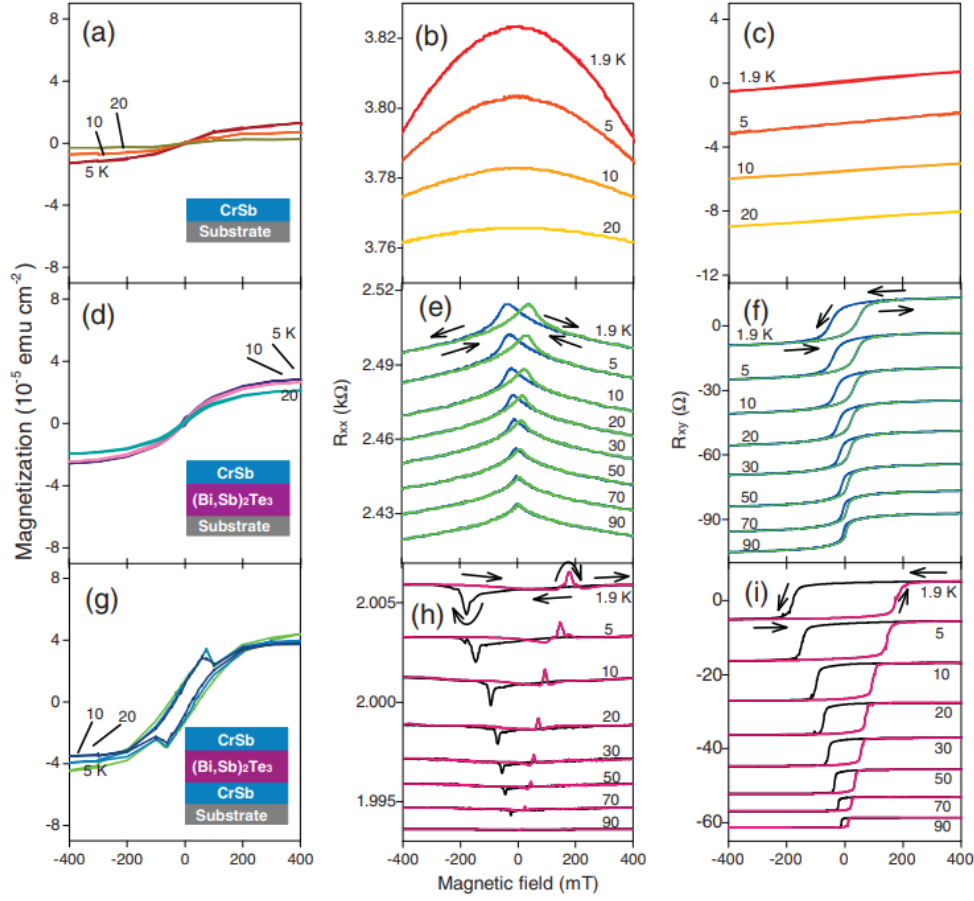


Figure 6 (a) Temperature-dependent M-H loops of a 12 nm AFM thin film, CrSb, grown on a GaAs substrate, which exhibits negligible magnetization. (b) and (c) show the results of the longitudinal ( $R_{xx}$ ) and the Hall ( $R_{xy}$ ) resistances, respectively. The absence of hysteresis in both measurement results, i.e., the small parabolic curvature in  $R_{xx}$  and the linear response in  $R_{xy}$ , suggests that the CrSb layer does not generate an AHE intrinsically. (d)–(f) show the corresponding results of an AFM/TI bilayer, demonstrating the transport signature of the top-surface magnetization of the TI layer. (g)–(i) are from an AFM/TI/AFM trilayer, in which an antisymmetric MR behavior and an AHE are observed.